Observation of the spontaneous vortex phase in the weakly ferromagnetic superconductor $ErNi_2B_2C$: A penetration depth study

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The coexistence of weak ferromagnetism and superconductivity in $ErNi_2B_2C$ suggests the possibility of a spontaneous vortex phase (SVP) in which vortices appear in the absence of an external field. We report evidence for the long-sought SVP from the in-plane magnetic penetration depth $\Delta\lambda(T)$ of high-quality single crystals of $ErNi_2B_2C$. In addition to expected features at the Néel temperature $T_N = 6.0$ K and weak ferromagnetic onset at $T_{WFM} = 2.3$ K, $\Delta\lambda(T)$ rises to a maximum at $T_m = 0.45$ K before dropping sharply down to ∼0.1 K. We assign the 0.45 K-maximum to the proliferation and freezing of spontaneous vortices. A model proposed by Koshelev and Vinokur explains the increasing $\Delta\lambda(T)$ as a consequence of increasing vortex density, and its subsequent decrease below T_m as defect pinning suppresses vortex hopping.

It is now clear that the borocarbide superconductor $ErNi_2B_2C$ develops weak ferromagnetism (WFM) below $T_{WFM} = 2.3$ K while remaining a singlet superconductor [1, 2]. The question naturally arises: how do these two seemingly incompatible orders — ferromagnetism and superconductivity — coexist microscopically? Clearly superconductivity will be suppressed if the internal field B_{in} generated by the ferromagnetic moment exceeds H_c for a Type-I, or H_{c2} for a Type-II, superconductor (SC). For a Type-II SC, however, vortices are predicted to appear *spontaneously* if B_{in} lies in the range $H_{c1} < B_{in} \sim 4\pi M < H_{c2}$ [3, 4, 5, 6]. In this spontaneous vortex phase (SVP), the vortex screening currents shield superconducting regions from the intrinsic magnetization. The vortices, however, may be qualitatively different from those generated by externally applied fields [7]. In this Letter we report unusual features in the penetration depth data of a high quality single-crystal of $ErNi_2B_2C$ that give strong evidence for the existence of the SVP.

There have been previous SVP reports that we consider inconclusive. Ng and Varma [8], for example, interpreted small angle neutron scattering (SANS) data on $ErNi_2B_2C$ as a prelude to the SVP. In that experiment, Yaron et al. [9] reported that the vortex-line lattice begins to tilt away from the c-axis (along which the magnetic is field applied) towards the a-b plane below T_{WFM}. However, the tilt can merely be a result of the vector sum of the applied field and the internal field produced by the ferromagnetic domains in the basal plane. Additional evidence was provided by SANS data [10] with the applied magnetic field in the basal plane. A large field was applied to align ferromagnetic domains. When the field was removed, the flux line lattice was found to persist below T_{WFM} but disappear above it. However, owing to the low T_c (∼ 8.5 K) and increased pinning below T_{WFM} [11], trapped flux cannot be ruled out.

Among the magnetic members of the rare-earth (RE) nickel borocarbide family, $RENi_2B_2C$ (RE = Ho, Er, Dy, etc.), $ErNi_2B_2C$, is a particularly good candidate for study. Superconductivity arises at $T_c \approx$ 11 K and persists when antiferromagnetic (AF) order sets in at $T_N \approx 6$ K [12]. In the AF state the Er spins are directed along the b-axis, forming a transversely polarized, incommensurate sinusoidal spin-density-wave (SDW) state, with modulation vector modulation vector $\delta = 0.553a^*$ ($a^* = 2\pi/a$) [13]. The appearance of higher-order reflections at lower temperatures [1] signals the development of a square-wave modulation, with regular spin slips spaced by $20a$. Below 2.3 K WFM appears with $B_{in} \cong 0.1$ T, approximately one Er magnetic moment per twenty unit cells, clearly correlated with spin slips.

The relative stability of various phases of a ferromagnetic superconductor was explored [5] by Greenside *et al.* A spiral phase is not possible in the presence of strong uniaxial anisotropy and the spontaneous vortex phase is more stable than a linearly polarized state for small values of $\zeta = [F_{FM}/F_s]$, the ratio of ferromagnetic to superconducting free-energy densities at $T = 0$. For $\zeta = 100$ and the ratio $\lambda/\gamma = 10$, where

$\gamma = [3k_BT_cS/(2aM^2(S+1)]^{1/2}$ is related to the exchange stiffness, Greenside *et al.* find the SVP to be the most stable low-temperature phase; indeed, they suggest that the effect is most likely to be found in a dilute ferromagnetic superconductor, and that smaller values of ζ favor SVP. In the case of $ErNi_2B_2C$, where only 5% of the Er atoms contribute to ferromagnetism, we have $F_s = -H_c^2/8\pi \approx -1.5 \times 10^5$ erg/cm^3, where $H_c \approx$ 1900 G from Ref. 2. The ferromagnetic energy density is $F_{FM} = -3Nk_BT_cS/(2(S+1)) \approx -4.3 \times 10^6$ erg/cm^3, where N=1.5x10^{22} cm^{-3} is the density of the (magnetic) Er atoms, and $S = 3/2$ is the Er spin. This then gives $\zeta = 30$, strongly favoring the SVP. The spin-stiffness length is $\gamma = 100$ Å at low temperatures where $M \approx 88$ G, so that $\lambda/\gamma \approx 7$, close to the value assumed in Ref. 5. As $ErNi_2B_2C$ is strongly Type II ($\lambda/\xi \approx 5$), we conclude that the SVP phase is the preferred state for coexisting ferromagnetism and superconductivity.

We have measured the temperature dependence of the in-plane magnetic penetration depth $\Delta\lambda(T) = \lambda(T) - \lambda(T_{base})$, in single crystals of $ErNi_2B_2C$ down to $T_{base} =$ 0.12 K using a tunnel-diode based, self-inductive technique at 21 MHz [14] with a noise level of 2 parts in 10^9 and low drift. The magnitude of the ac field was estimated to be less than 40 mOe. The cryostat was surrounded by a bilayer Mumetal shield that reduced the dc field to less than 1 mOe. *The very small values of the ac and dc field in our system ensure that our measurement is essentially a zero-field one, thereby eliminating the possibility of trapped flux.* Details of sample growth and characterization are described in Ref. 12. The samples were then annealed according to conditions described in Ref. 15. The sample was mounted, using a small amount of GE varnish, on a single crystal sapphire rod. The other end of the rod was thermally connected to the mixing chamber of an Oxford Kelvinox 25 dilution refrigerator. The sample temperature is monitored using a calibrated RuO_2 resistor at low temperatures (T_{base} – 1.8 K), and a calibrated Cernox thermometer at higher temperatures (1.3 K – 12 K).

The deviation $\Delta\lambda(T) = \lambda(T) - \lambda(0.12\text{ K})$ is proportional to the change in resonant frequency $\Delta f(T)$ of the oscillator, with the proportionality factor G dependent on sample and coil geometries. We determine G for a pure Al single crystal by fitting the Al data to extreme nonlocal expressions and then adjust for relative sample dimensions [16]. Testing this approach on a single crystal of Pb, we found good agreement with conventional BCS expressions. The value of G obtained this way has an uncertainty of ±10% because our sample, with approximate dimensions $1.2 \times 0.9 \times 0.4$ mm^3, has a rectangular, rather than square, basal area [17].

Figure 1 shows the temperature-dependence of the in-plane penetration depth $\Delta\lambda(T)$. We see the following features: (1) onset of superconductivity at $T_c^* = 11.3$ K, (2) a slight shoulder at $T_N = 6.0$ K, (3) a broad peak at

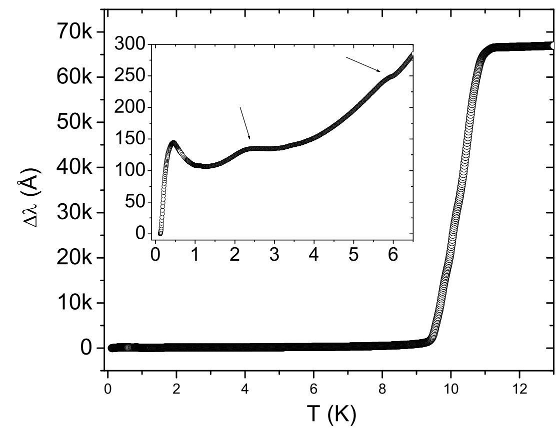

FIG. 1: Temperature dependence of the penetration depth $\Delta lambda(T)$ from 0.12 K to 13 K. Inset shows the low-temperature region. The arrows point to the features at T_N and T_{WFM}.

$T_{WFM} = 2.3$ K, (4) another sharp peak at $T_m = 0.45$ K, and (5) an eventual downturn below T_m. The features at T_N and T_{WFM} have not been seen in previous microwave measurements of $\Delta\lambda(T)$ on either thin-film [18] or single-crystal $ErNi_2B_2C$ [19] but the former has been observed in SANS data [20]. We show in a separate publication [21] that the feature at T_N is only observed for relatively small non-magnetic scattering rates. The large value of T_c and the resolvability of the features at T_N and T_{WFM} attest to the high purity of the samples. We show, for comparison, data for a sample grown by floating-zone methods in the inset to Fig. 2. No clear signal is seen at T_N, although there may be some sign of the Neel transition near 5 K. In place of the up-turn in the penetration depth, the signal levels off near T_{WFM} before decreasing below 1 K. This suggests that spontaneous vortices at the surface of this sample are strongly pinned.

Figure 2 shows the data below T_{WFM}. The strong upturn is a significant deviation from the normal monotonic decrease of the penetration depth with decreasing temperature. Because we expect the Meissner effect to vanish ($\lambda(T)$ to diverge) in the SVP in the absence of pinning [8], it is natural to analyze the low-temperature data in the context of weakly pinned vortices in the low-frequency limit. We use a two-level tunneling model proposed by Koshelev and Vinokur (KV) [22]. This approach has been revisited by Korshunov [23] and applied to ultrathin cuprate films by Calame *et al.* [24]. At relatively high frequencies, small oscillations of the pinned lattice near equilibrium (Campbell regime) dominate absorption. At lower frequencies, jumps of lattice regions between different metastable states (two-level systems) come into play and determine the absorption. Both regimes are sensitive to the pinning strength, which depends on the

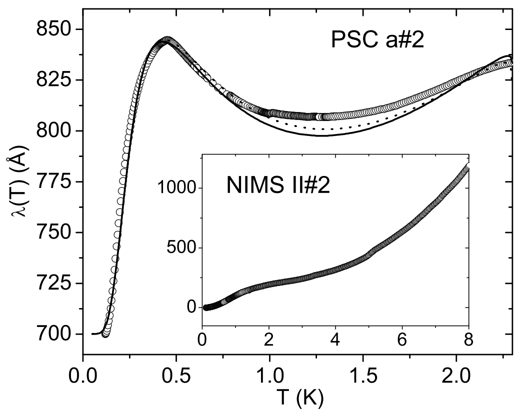

FIG. 2: (◯) Temperature dependence of the penetration depth $\Delta\lambda(T)$ in the WFM phase. We've assumed $\lambda(T_{base}) \approx \lambda(0)$=700 Å from Ref. 20. The solid line is the fit of the data to Eqn. 1 for n=0.21, while the dotted line is that of n=0.125. The values of other parameters are mentioned in the text. The inset shows $\Delta\lambda$ for a floating-zone-grown sample exhibiting no signals at T_N or T_{WFM}

value of the internal magnetic field B_{in}. At small field, vortices are pinned independent of one another. When B_{in} exceeds a characteristic value B_p, pinning becomes collective [25], and the vortex lattice splits into volumes that are pinned as a whole, with correlated regions having length L_{c0} parallel to the field. We argue below that, in the WFM phase, $H_{c1} < B_{in}, B_p \ll H_{c2}$ and that the frequency lies in the two-level regime.

In the two-level model, the pinning state is characterized by a large number of neighboring metastable configurations. If we shift any volume V of the vortex lattice, then a finite probability exists that at some distance u there is another state with an energy within Δ of the starting configuration and separated from it by a barrier U. At finite temperatures such regions of the lattice jump among metastable states. Under the action of an applied ac field of frequency ω, the ac screening current exerts an ac Lorentz force on the vortex lattice which induces jumps among nearby metastable states. The motion of the vortices, which carry magnetic fields, then increases the ac field penetration into the sample, resulting in an increase in the effective penetration depth λ_{eff}. The system exhibits typical Debye-type behavior with the properties determined by $\tau(T) \sim \tau_0 \exp(U/T)$, the mean time between jumps. When two-level response dominates over Cambell behavior the penetration is estimated [22] to behave as

$$\lambda_{eff}^2(T) = \lambda_{\parallel}^2(T) + \frac{B_{in}^2(T) n_{tl}}{16\pi T} \left\langle \frac{V^2 u^2}{(1+(\omega\tau(T))^2)\cosh^2(\Delta/T)} \right\rangle \tag{1}$$

where $\langle ... \rangle$ denotes an average over the distribution of two level systems. Here $\lambda_{\parallel}(T)$ is the London penetration depth in the absence of vortices; B_{in}, the internal magnetic field; and n_{tl}, the concentration of two-level systems. In the low-field region ($B_{in} < B_p$), the vortex lines move independently, and their presence does not change the penetration depth considerably ($\lambda_{eff} \approx \lambda_{\parallel}$). However, in the collective pinning state ($B_{in} > B_p$), the jumping volume is not too small, and the characteristic distance at which the nearest metastable state exists is approximately the radius of the pinning force $u \approx \xi_{\parallel}$. As the temperature decreases, there is insufficient thermal energy to overcome the barrier U. No jumping takes place, the vortices are frozen, and hence there is no extra penetration. One therefore recovers the London penetration depth $\lambda_{\parallel}$ at the lowest temperatures.

The solid line shows the fit of Eq. 1 to the data below T_{WFM}. In this fit, we follow KV and replace $\langle ... \rangle$ with values that characterize an effective number n_{tl}^{eff} of active two-level systems. The values of the following quantities will be justified later: $B(T=0) \approx 1100$ G, $u \approx \xi_{\parallel} \approx 150$ Å, $V = L_{c0}u^2 = 5.4 \times 10^{-16}$ cm^3, and $\tau_0 = 2.2 \times 10^{-9}$ s. The temperature-dependence of the internal magnetic field $B(T)$ can be obtained by fitting magnetization values in Ref. 26 to the expression

$$B(T) \sim \left(1 - \frac{T}{T_{WFM}}\right)^n \tag{2}$$

giving n=0.21. This value of n is between the 2D-Ising value of 0.125 and the 3D-Ising value of 0.31, which is reasonable because in $ErNi_2B_2C$ the spins lie on sheets normal to the a axis and are confined to be along or anti-along the b axis, yet there is also 3D behavior in the superconductivity. Because U and Δ are strongly correlated, we make the reasonable assumption that all metastable states are equivalent ($\Delta = 0$) and choose the energy barrier $U = 0.49$ K and pinning density $n_{tl}^{eff} = 1.61 \times 10^{11}$ cm^{-3} that best fit the peak in $\lambda_{eff}(T)$. This value of the barrier makes $\omega\tau \approx 1$ near 1 K. Note that this value of U is close to the position of the peak at T_m — this is reasonable since below this temperature, the vortices no longer have enough thermal energy to overcome the barrier to hop among metastable states; hence, one recovers the Meissner state with λ decreasing. We expect $\lambda_{\parallel}(T)$ to exhibit a power-law temperature dependence at low temperatures from the combination of gap-minima observed in non-magnetic borocarbides and the increased pair-breaking as Er spins disorder. Consequently, we set $\lambda_{\parallel}(T) = \lambda_{\parallel}(0)(1 + bT^2)$ with $b = 0.036$ K^{-2} the third adjustable parameter in the fit. For comparison, we show the (dotted-line) fit with $n = 1/8$ (2D-Ising model) for which $U = 0.49$ K, $n_{tl}^{eff} = 1.57 \times 10^{11}$ cm^{-3}, and $b = 0.033$ K^{-2}. Both fits reproduced the qualitative features of the data, though the latter curve fits the data slightly better.

To justify our application of the two-level model to our data, we evaluate various physical parameters in

Quantity	Expression	Value	Notes
Depairing current, j_s	$c\Phi_0/(12\sqrt{3}\pi^2\xi_\parallel\lambda_\parallel^2)$	1.5×10^8 A/cm^2	
Viscous drag coefficient, η	$\approx \Phi_0^2/(2\pi\xi_\parallel^2\rho_n c^2)$	5.6×10^{-7} erg s cm^{-3}	Bardeen-Stephens model
Bean-model critical current, j_c	$4cM_h/L$	1.9×10^4 A/cm^2	Ref. [11], $L=1$ mm, M_h from hysteresis loop
Flux coherence length, L_{c0}	$x^2=\frac{j_s}{j_c}\ln x$; $x=\frac{\lambda_\perp L_{c0}}{\lambda_\parallel\xi_\parallel}$	19.5 nm	Ref. [12],[22], $\lambda_\perp/\lambda_\parallel\approx1.3$
Jump time prefactor, τ_0	$\eta\xi_\parallel c/(\Phi_0 j_c)$	2.2×10^{-9} s	Ref. [22]
Collective pinning field, B_p	$\Phi_0(\ln x)^{4/3}/L_{c0}^2$	20 Oe	Ref. [22]
Campbell crossover, $\omega_{cr}(B,T)$	$\approx T\Phi_0/(\eta BV\xi^2)$	27 MHz	Ref. [22], B=500 Oe; T=2 K

TABLE I: Vortex parameters for the two-level hopping regime

the model using standard expressions for the vortex state [27]. We start with the measured quantities: zero-temperature in-plane penetration depth $\lambda_\parallel(0)\approx$ 700 Å and coherence length $\xi_\parallel\approx150$ Å (and $\kappa=\lambda_\parallel(0)/\xi_\parallel=4.7$) from Ref. 20, ferromagnetic moment $M\approx0.62\mu_B$/Er $\approx$ 100 Oe well below T_{WFM} from Ref. 11, and the normal-state resistivity $\rho_n(T_c^*)=5.8\ \mu\Omega$ cm that we measured for our sample. The value of M corresponds to an internal field $B\sim4\pi M=1100$ G, which is greater than $H_{c1}\sim500$ G, putting us in the mixed state.

In Table I, we give the expressions and values for the quantities that lead to the flux coherence length L_{co} (19.5 nm), the collective pinning field B_p (20 Oe), the frequency ω_{cr} above which Campbell response is expected (27 MHz), and the jump-time prefactor τ_0 (2.2 ns). Since we operate at 21 MHz, this puts us in the two-level regime. Note that B_p is less than H$_{c1}$, suggesting the the mixed state of $ErNi_2B_2C$ is always in the collective pinning regime. Based on these values, we estimate the maximum density of two-level volumes to be $\sim2.4\times10^{13}$cm^{-3} at the lowest temperatures, indicating that approximately 1% are active in our frequency window.

KV [22] also found the Campbell penetration depth to be $\sim B^2$, which is a monotonically increasing function with decreasing temperature, i.e. there is no peak at low temperatures. This is in agreement with our not being in the Campbell regime. We also measured $\lambda(T)$ with the ac field along the basal plane, finding features qualitatively similar to the present data, including the strength and position of the features at T_N, T_{WFM} and T_m.

In conclusion, penetration depth data of single-crystal $ErNi_2B_2C$ down to $\sim$0.1 K provide strong evidence for the existence of a spontaneous vortex phase below T_{WFM}. The high quality of our sample enables us to see features at T_N and T_{WFM} that have not been observed in previous studies of the penetration depth [18, 19]. Other samples, such as that shown in the inset to Fig. 2 show no clear signal at either T_N or T_{WFM}, nor the upturn in penetration depth that we attribute to weakly pinned spontaneous vortices. As pointed out by Radzhiovsky [7], the SVP lattice is much softer than a conventional lattice, and therefore especially sensitive to quenched disorder. It may well be that the spontaneous vortices may be glass-like rather than forming a lattice, and that ferromagnetic closure domains form at the surface. While these aspects may make spontaneous vortices difficult to detect by neutron scattering or surface magnetization, vortices will still have strong effects on the electrodynamics, as observed here.

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[1] S.-M. Choi *et al.*, Phys. Rev. Lett. **87**, 107001 (2001).
[2] P. C. Canfield, S. L. Bud'ko, and B. K. Cho, Physica C **262**, 249 (1996).
[3] E. I. Blount and C. M. Varma, Phys. Rev. Lett. **42**, 1079 (1979).
[4] M. Tachiki, A. Kotani, H. Matsumoto, and H. Umezawa, Solid State Commun. **31**, 927 (1979).
[5] H. S. Greenside, E. I. Blount, and C. M. Varma, Phys. Rev. Lett. **46**, 49 (1981).
[6] C. G. Kuper, M. Revzen, and A. Ron, Phys. Rev. Lett. **44**, 1545 (1980).
[7] L. Radzihovsky, A. M. Ettouhami, K. Saunders, and J. Toner, Phys. Rev. Lett. **87**, 027001 (2001).

[8] T. K. Ng and C. M. Varma, Phys. Rev. Lett. **78**, 330 (1997).
[9] U. Yaron *et al.*, Nature (London) **382**, 236 (1996).
[10] H. Kawano-Furukawa, H. Yoshizawa, H. Takeya, and K. Kadowaki, J. Magn. Magn. Mater. **226-230**, 278 (2001).
[11] P. L. Gammel *et al.*, Phys. Rev. Lett. **84**, 2497 (2000).
[12] B. K. Cho *et al.*, Phys. Rev. B **52**, 3684 (1995).
[13] J. Zarestky *et al.*, Phys. Rev. B **51**, R678 (1995).
[14] I. Bonalde *et al.*, Phys. Rev. Lett. **85**, 4775 (2000).
[15] X. Y. Miao, S. L. Bud'ko, and P. C. Canfield, J. Alloys. Comp. **338**, 13 (2002).
[16] E. E. M. Chia *et al.*, Phys. Rev. B **67**, 014527 (2003).
[17] R. Prozorov, R. W. Giannetta, A. Carrington, and F. M. Araujo-Moreira, Phys. Rev. B **62**, 115 (2000).
[18] A. Andreone *et al.*, Physica C **319**, 141 (1999).
[19] T. Jacobs *et al.*, Phys. Rev. B **52**, R7022 (1995).
[20] P. L. Gammel *et al.*, Phys. Rev. Lett. **82**, 1756 (1999).
[21] E. E. M. Chia *et al.*, unpublished .
[22] A. E. Koshelev and V. M. Vinokur, Physica C **173**, 465 (1991).
[23] S. E. Korshunov, Phys. Rev. B **63**, 174514 (2001).
[24] M. Calame, S. E. Korshunov, C. Leemann, and P. Martinoli, Phys. Rev. Lett. **86**, 3630 (2001).
[25] A. I. Larkin and Y. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).
[26] J. Jensen, Phys. Rev. B **65**, 140514(R) (2002).
[27] G. Blatter *et al.*, Rev. Mod. Phys. **66**, 1125 (1994).